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Uniaxial Stress Study of the Magnetotransport Properties of the quasi-two Dimensional Organic Conductor α-(BEDT-TTF)$_2$KHg(SCN)$_4$

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Abstract

Measurements of the low temperature magnetoresistance of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ under uniaxial stress perpendicular to the BEDT-TTF molecular layers indicate that the density wave ground state is suppressed by 1.5 Kbar. The fundamental Shubnikov-de Haas frequency decreases at an approximate rate of -20 tesla/Kbar and a slow, stress dependent oscillation appears for stresses above 0.9 Kbar. These enhanced stress effects are discussed in light of the high sensitivity of the Fermi surface to small changes in the crystal structure.

INTRODUCTION

Hydrostatic pressure studies of the transport properties of the α-(BEDT-TTF)$_2$MHg(SCN)$_4$ (M = NH_4, K, Tl, or Rb) family of quasi-two dimensional charge transfer organic conductors [1] have revealed that the salts are extremely sensitive to small changes in the lattice parameters [2,3]. In α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$, the superconducting ground state (T_c = 1 K) is suppressed with only a few Kbar of hydrostatic pressure and a low frequency (≈ 48 tesla) pressure sensitive oscillation superposed on the linear magnetoresistance (MR) background starts appearing at about 2 Kbar [4]. These two effects have been attributed to imperfect nesting of the electron open orbits [5], which suppresses the superconductivity and creates a small pocket on the Fermi surface. Hydrostatic pressure applied to the other members of the family, M=K, Tl, and Rb, destroys the density wave ground state present in these salts at temperatures below 8, 10, and 11 K respectively. This is evidenced by a reduction of the giant MR anomaly and the disappearance of the kink field (H_k), which is believed to mark the normal state/density wave magnetic field boundary [2].

In simple terms, the application of hydrostatic pressure to a single crystal *reduces* the lengths of all three lattice vectors, while uniaxial stress *contracts* the lattice vector in the direction of the applied stress and, due to Poisson's effect, *expands* the lattice parameters along the plane perpendicular to the applied stress. In the case of layered molecular crystals, uniaxial stress applied perpendicular to the molecular planes is of obvious interest since it produces a decrease in the inter-layer spacing accompanied by an increase in the intra-layer dimensions, a configuration unreachable with hydrostatic pressure. The effects of tensile stress along the BEDT-TTF molecular planes [6] and low compressive stress perpendicular to the planes [7] on the superconductivity of (BEDT-TTF)$_2$Cu(NCS)$_2$ have been studied using different methods [8]. We have used a new sample preparation technique to study the low temperature MR of the title compound, α-(BEDT-TTF)$_2$KHg(SCN)$_4$, under high uniaxial stress along the b-axis, i.e. perpendicular to the planes of the BEDT-TTF molecules. The basic idea is to compensate for the crystal's fragility and surface irregularities by encapsulating it in a medium with closely matched physical properties and whose shape can be optimized to sustain the applied stress.

EXPERIMENTAL

Four gold wires were bonded with gold paint to the plate-like faces of a small (≈ 0.5×0.5 mm^2 faces) and thin (≤ 0.3 mm) single crystal of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ for the standard four terminal resistance measurement across the BEDT-TTF planes. The crystal was placed in a drop of liquid two-component epoxy between two sheets of kapton with its flat faces parallel to the sheets. A small, solid cylindrical crystal-epoxy tube was obtained after the epoxy cured. Stress was then applied on the flat faces

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of the tube, that is, along the crystal's b-axis, using a pneumatic piston ^{3}He insert. A more detailed description of the sample preparation method and the stress apparatus can be found in Ref. [9]. The resistance was measured by AC and DC techniques with currents in the range 10-100 μA. Magnetoresistance data for various single crystals were taken in resistive and hybrid magnets at the High Field Magnet Laboratory in the University of Nijmegen and at the Francis Bitter National Magnet Laboratory in MIT, Cambridge.

RESULTS AND DISCUSSION

Figure 1 shows the MR of a crystal of α-(BEDT-TTF)$_2$KHg(SCN)$_4$ (sample A) at 0.55 K for increasing values of b-axis stress (P_b). At zero stress (no gas pressure in the piston) all the MR characteristics observed in atmospheric pressure crystals are present: the anomalous giant MR that peaks off around 14 tesla, followed by a region of negative MR and a sharp drop around 23.5 tesla (the kink field), and the strong Shubnikov-de Haas (SdH) oscillations superposed on the background. In the region below H_k a second peak in the SdH signal is faintly visible, whereas above H_k only a single large amplitude oscillation is present. As the stress is progressively increased, the MR peak reduces continuously and H_k moves down to lower field values. For stresses above 1 Kbar, these two features become intermingled with a slow oscillation riding on the background MR (more on this oscillation later) and it becomes hard to identify the sharp drop in MR that is a mark of the kink transition. However, the transition can also be identified with a strong field hysteresis between the up and down field sweep values where the drop occurs. With this in mind, the kink field can be seen to slowly vanish as the stress approaches 1.5 Kbar (see Fig. 2). The disappearance of the kink field can perhaps be best seen in Fig. 3, where the MR data for a different α-(BEDT-TTF)$_2$KHg(SCN)$_4$ single crystal (sample B) are shown. For this sample, the high magnetic field MR becomes flat for stress values above 0.9 Kbar and, for P_b = 2.2 Kbar it is actually *increasing* at 24 tesla. Clearly, the density wave ground state in this salt is being suppressed with increasing uniaxial stress. This response is similar to the behavior observed under hydrostatic pressure, but occurs at a much lower value of absolute pressure (≈ 1.5 Kbar as opposed to 5 Kbar) [2,3].

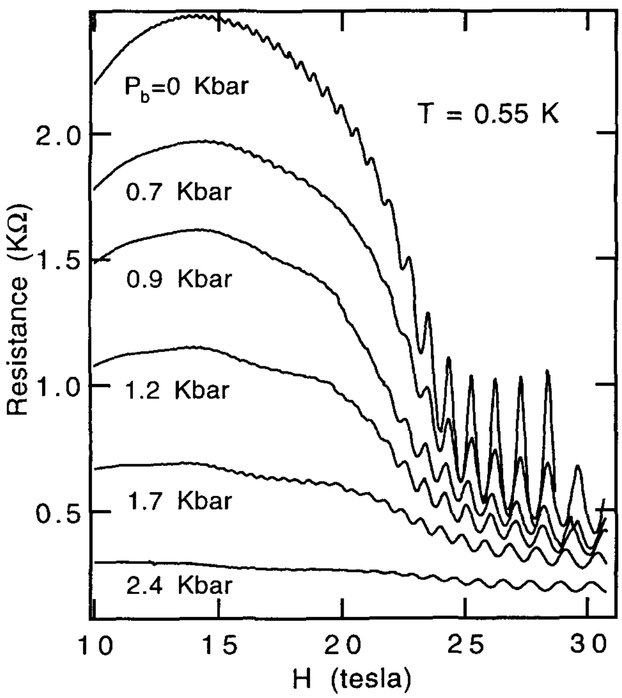

Figure 1. MR of (sample A) measured in a hybrid 30 tesla magnet for increasing values of uniaxial stress applied along the crystal's b-axis (only the up magnetic field sweeps are shown).

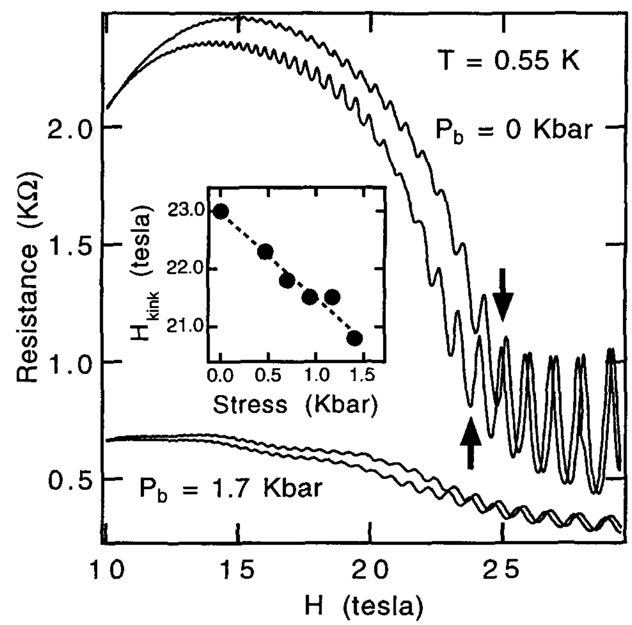

Figure 2. Up and down magnetic field sweeps MR for P_b = 0 and 1.7 Kbar. Arrows indicate the position of the kink field at 0 stress for each sweep. Note the absence of field hysteresis for the 1.7 Kbar curve. Inset: Stress dependence of the kink field (as measured in the down field sweeps).

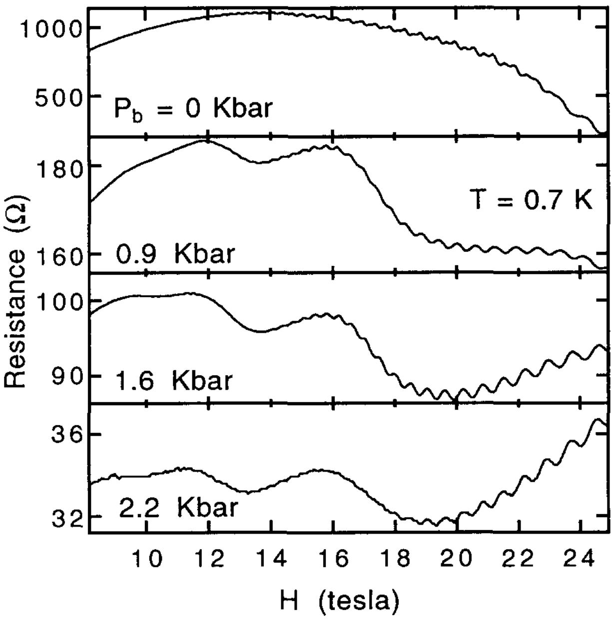

Figure 3. Magnetoresistance of sample B at 0.7 K for different values of b-axis stress. Maxima and minima of the slow oscillation are evident.

Shubnikov-de Haas oscillations in the MR of both samples are clearly visible for all stresses (Figs. 1 and 3) and Fourier spectral analysis of the signals reveals that the fundamental SdH frequency (α) decreases with stress from a zero stress value of 668±6 tesla (the accepted atmospheric pressure value is 670 tesla [2]) at a rate of -20±4 tesla/Kbar [10] (Fig. 4). The decrease in α can be attributed to a decrease in the size of the first Brillouin zone (BZ), which is caused by the expansion of the a-c plane unit cell due to Poisson's effect. [It is important to contrast this with the effect of hydrostatic pressure, which increases α by compressing the unit cell (see Fig. 4).] However, if attributed solely to the contraction of the BZ, the rate of change of α corresponds to an increase in the a-c plane unit cell area of 0.03/Kbar. This value is almost an order of magnitude higher than what is expected for the compressibility of this salt [11]. Thus, most probably, adjustments in the relative positions of the BEDT-TTF molecules take place, which modify the shape of the Fermi Surface (FS) via shifts in the molecular overlap integrals and contribute to the decrease in the relative size of the closed orbits.

It is also these rearrangements in overlap integrals that are most likely to cause the appearance of a slow oscillation in the MR for stresses above 0.9 Kbar. Figure 3 shows the detail of these oscillations in sample B for various stress values. Similar stress induced oscillations can be

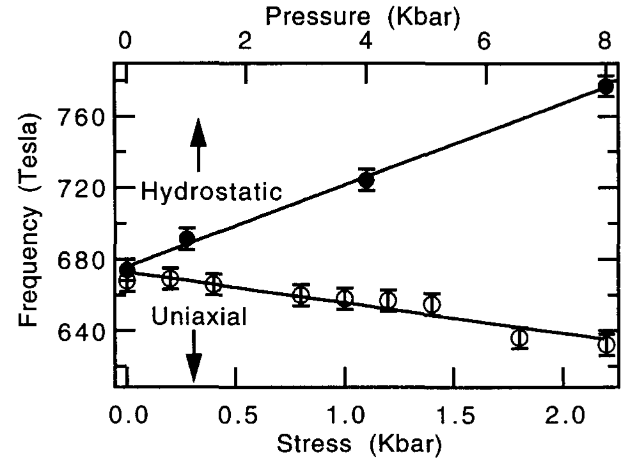

Figure 4. Variation of the fundamental SdH frequency with hydrostatic pressure (top scale, data from Ref. [2]) and uniaxial stress (bottom scale). Note the difference in pressure range between the top and bottom scales.

seen in sample A (Fig. 1) and in all the other samples we have measured (not shown). After proper subtraction of the faster SdH signal and the slowly changing MR background, the oscillation maxima and minima can be indexed with integer and half-integer numbers. A plot of the indices versus inverse magnetic field reveals a frequency of approximately 52 tesla for the 0.9 Kbar oscillation, which corresponds to an extremal area of 1.3% of the atmospheric pressure BZ. The frequency decreases linearly with stress at a rate of -9.4 tesla/Kbar [13] (Fig. 5) and corresponds to a Landau level index equal to 2, very close to the quantum limit. As has already been mentioned, a similar oscillation has been observed in the superconducting sister compound α-$(BEDT\text{-}TTF)_2NH_4Hg(SCN)_4$ under hydrostatic pressure [4], which was attributed to imperfect nesting of the open

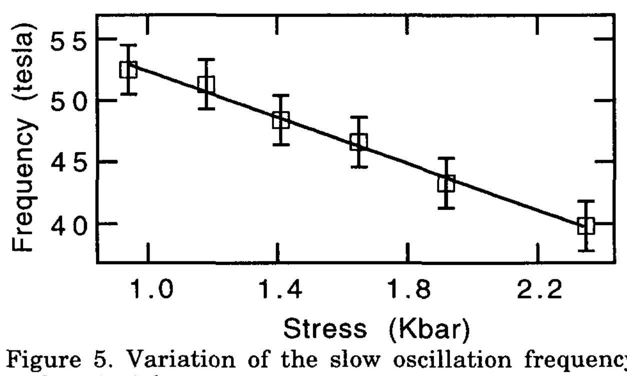

Figure 5. Variation of the slow oscillation frequency with uniaxial stress.

orbits. In the case of α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$, uniaxial stress is probably destroying the perfect nesting of the closed orbits, thereby suppressing the density wave state and creating a small closed orbit responsible for the slow oscillation observed. Another possibility is that, once again, the shifts in molecular orbital overlaps induced by stress result in a FS where the open orbits have been replaced by small close orbits. Recently, Ducasse and Fritsch discussed the sensitivity of the molecular overlap integrals in this family of salts [14]. Small changes in the relative positions of the molecules might lead to drastic modifications in the FS. This non-linearity might perhaps explain why, of all members of the α-$(BEDT\text{-}TTF)_2MHg(SCN)_4$ family, only $M=NH_4$ goes superconducting. The answer lies in the small differences that exist between the salts' unit cell parameters, relative positions of the molecules in the planes, and shape distortions of each individual molecule [5]. It is then not hard to imagine that hydrostatic pressure and uniaxial stress (which for crystals with non-cubic symmetry generate shears besides the contractions and elongations) might induce the changes in the electronic structure required to make two salts with different atmospheric pressure ground states behave identically under pressure or stress.

CONCLUSION

In summary, we have demonstrated that uniaxial stress produces some magnetotransport effects in α-$(BEDT\text{-}TTF)_2KHg(SCN)_4$ consistent with previous hydrostatic pressure results (suppression of the density wave state, decrease in the fundamental SdH frequency) and other features previously unobserved (low frequency orbit). The dramatic changes observed in this family of salts are likely to be due to the high sensitivity of the molecular overlap integrals, and, therefore, of the FS, to small changes in their crystal structures. Measurements of the elastic constants of these salts, preferably at low temperatures, would greatly improve the interpretation of our results as well as those obtained with hydrostatic pressure. Work is in progress to expand the sample preparation technique to allow the application of uniaxial stress along the two other crystal axes.

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